Sonder les modes optiques avec des nanosources fluorescentes

Remi Carminati

Sonder les modes optiques avec des nanosources fluorescentes

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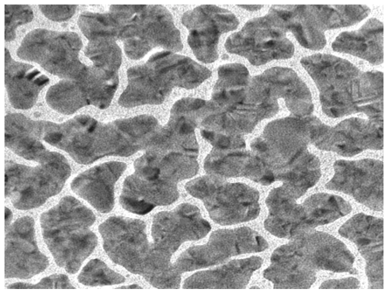

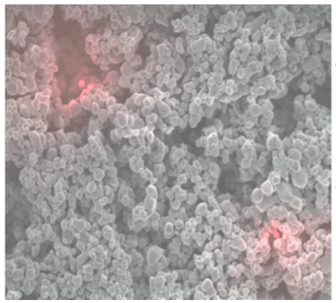

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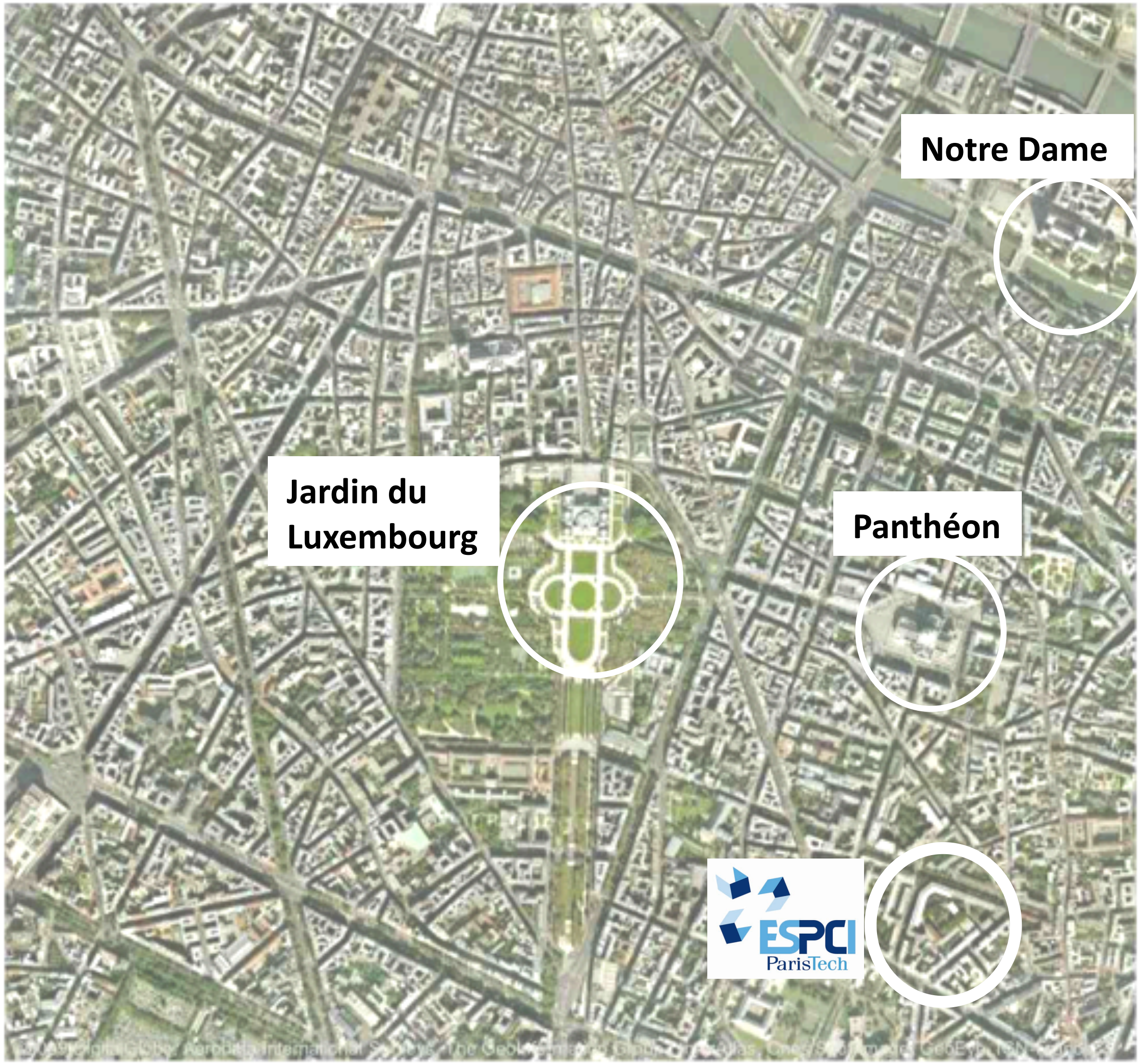
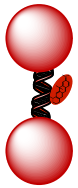
www.institut-langevin.espci.fr



Outline

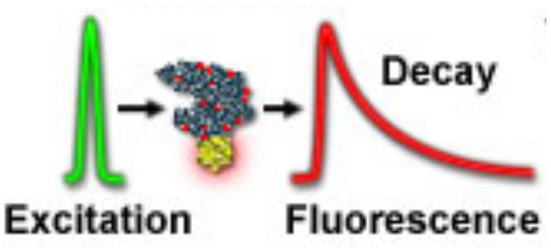


Spontaneous emission in nanostructured environments

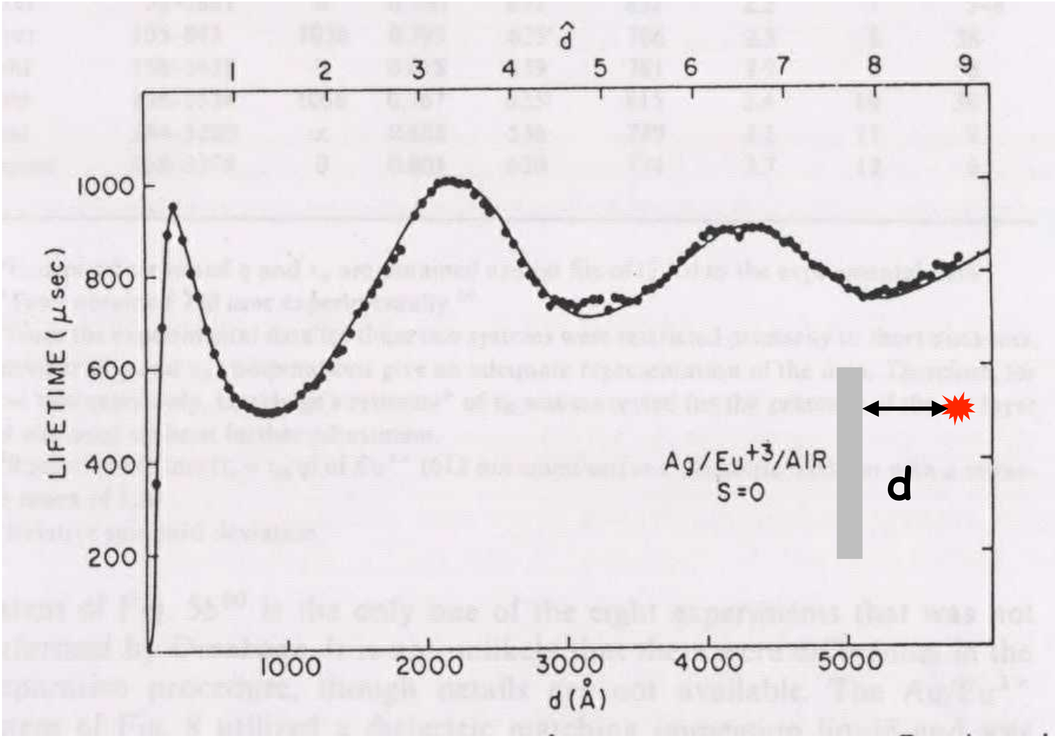
Probing localized plasmons on disordered metallic films

Probing near-field interactions in volume disordered systems (white powders)

Fluorescence depends on environment

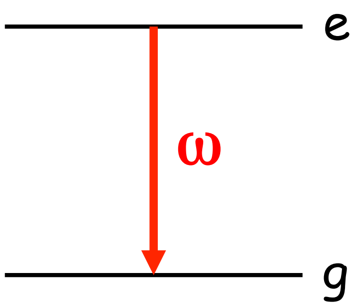


Lifetime close to a silver mirror



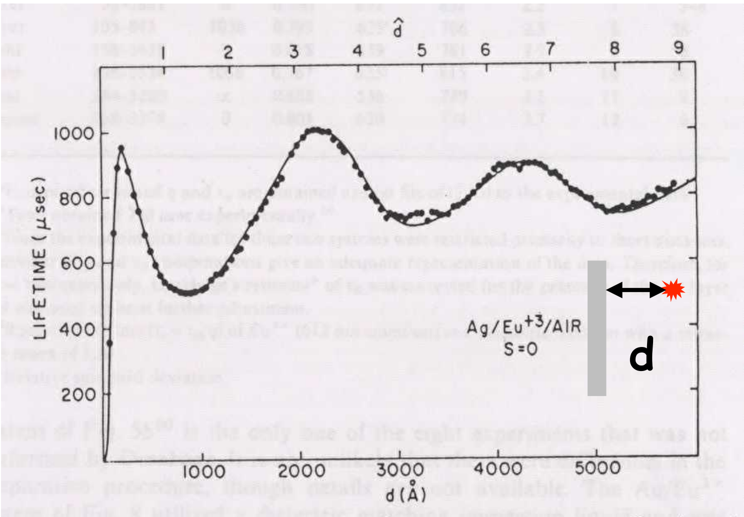
Drexhage (1970)
Chance, Prock, Silbey (1978)

How to describe the change in lifetime ?



Probability of being excited at time t $\quad P(t) \propto \exp(-\Gamma t)$

Lifetime of excited state $\quad \tau = 1/\Gamma$



Drexhage (1970)
Chance, Prock, Silbey (1978)

<u>Pertubation theory</u>
(Fermi golden rule)

$$\Gamma = \frac{\pi\omega}{3\varepsilon_0\hbar}\left|\mathbf{p}_{ge}\right|^2 \rho_{\mathbf{u}}(\mathbf{r}_0,\omega)$$

Local Density of States (LDOS)

$$\boxed{\frac{\Gamma}{\Gamma_0} = \text{change in the LDOS}}$$

Local Density Of States (LDOS)

- Density Of States (DOS)
  - Counts modes at a given frequency

$$\rho(\omega) = \sum_n \delta(\omega - \omega_n)$$

- Local Density Of States (LDOS)
  - Counts modes at a given frequency weighted by their contribution at point **r**

$$\rho(\omega, \mathbf{r}) = \sum_n |\mathbf{E}_n(\mathbf{r})|^2 \, \delta(\omega - \omega_n)$$

Purcell effect

The change in the LDOS describes the Purcell effect

E. M. Purcell "Spontaneous emission probabilities at radio frequencies" *Phys. Rev.* **69**, 681 (1946)

B10. Spontaneous Emission Probabilities at Radio Frequencies. E. M. PURCELL, *Harvard University.*—For nuclear magnetic moment transitions at radio frequencies the probability of spontaneous emission, computed from

$$A_\nu = (8\pi\nu^2/c^3)h\nu(8\pi^3\mu^2/3h^2)\ \mathrm{sec.}^{-1},$$

is so small that this process is not effective in bringing a spin system into thermal equilibrium with its surroundings. At 300°K, for $\nu=10^7$ sec.$^{-1}$, $\mu=1$ nuclear magneton, the corresponding relaxation time would be 5×10^{21} seconds! However, for a system coupled to a resonant electrical circuit, the factor $8\pi\nu^2/c^3$ no longer gives correctly the number of radiation oscillators per unit volume, in unit frequency range, there being now *one* oscillator in the frequency range ν/Q associated with the circuit. The spontaneous emission probability is thereby increased, and the relaxation time reduced, by a factor $f=3Q\lambda^3/4\pi^2V$, where V is the volume of the resonator. If a is a dimension characteristic of the circuit so that $V\sim a^3$, and if δ is the skin-depth at frequency ν, $f\sim\lambda^3/a^2\delta$. For a non-resonant circuit $f\sim\lambda^3/a^3$, and for $a<\delta$ it can be shown that $f\sim\lambda^3/a\delta^2$. If small metallic particles, of diameter 10^{-3} cm are mixed with a nuclear-magnetic medium at room temperature, spontaneous emission should establish thermal equilibrium in a time of the order of minutes, for $\nu=10^7$ sec.$^{-1}$.

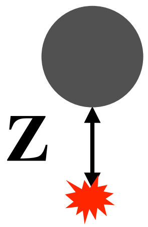
<u>For a single mode cavity</u>

$$\frac{\Gamma}{\Gamma_0} = \text{change in LDOS}$$

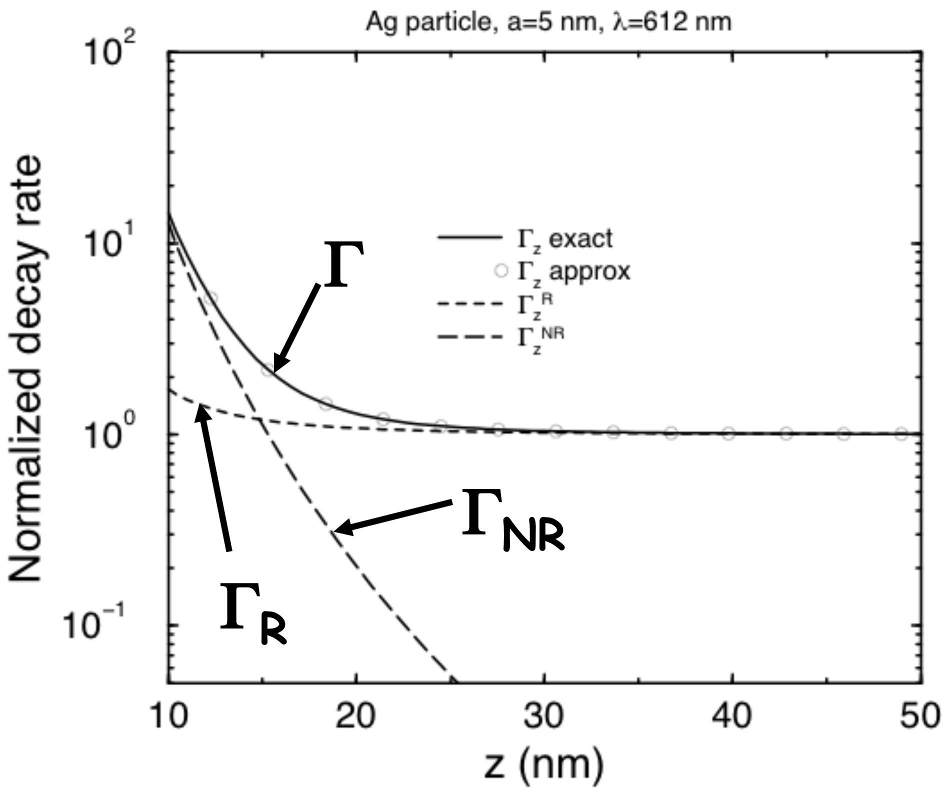
$$\frac{\Gamma}{\Gamma_0} = \frac{3}{4\pi^2}\lambda^3\frac{Q}{V}$$

(Purcell factor)

Interaction with a single nanoparticle



Silver nanoparticle
Diameter 10 nm



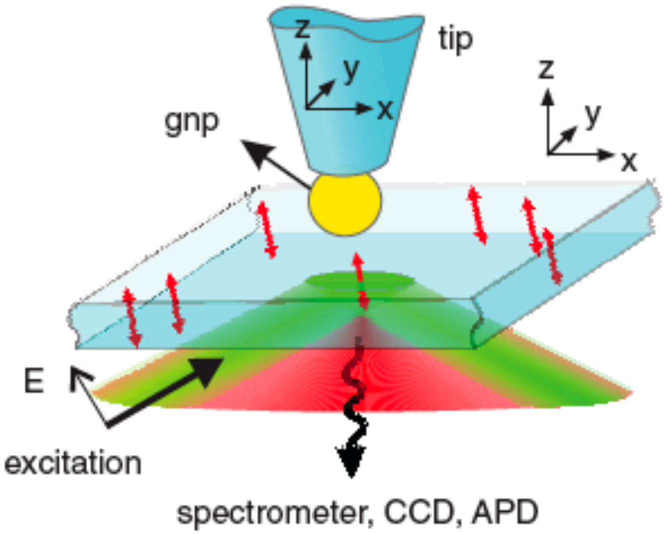
$$\Gamma = \Gamma_R + \Gamma_{NR}$$

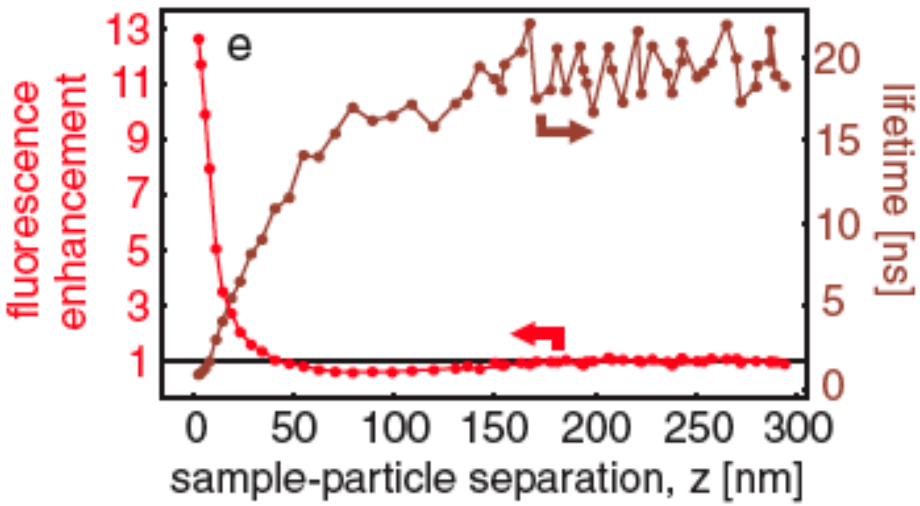
Photon emission

Absorption

Carminati *et al.*, Opt. Commun. **261**, 368 (2006)

Nanoscale controlled experiments on single emitter

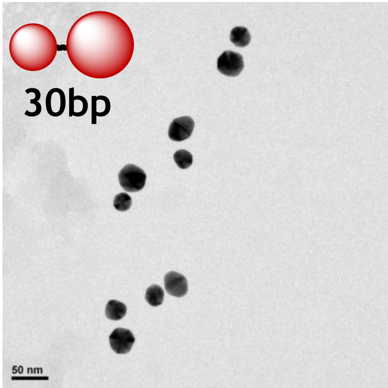


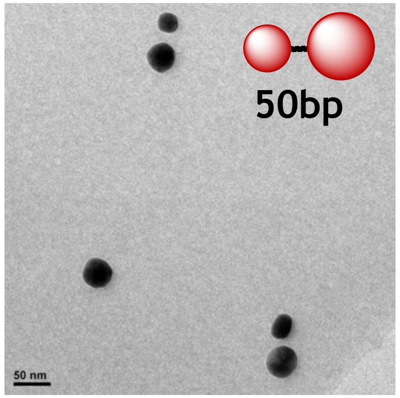


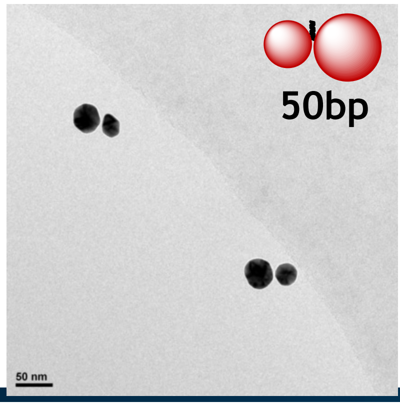
S. Kühn *et al.*, PRL **97**, 017402 (2006)

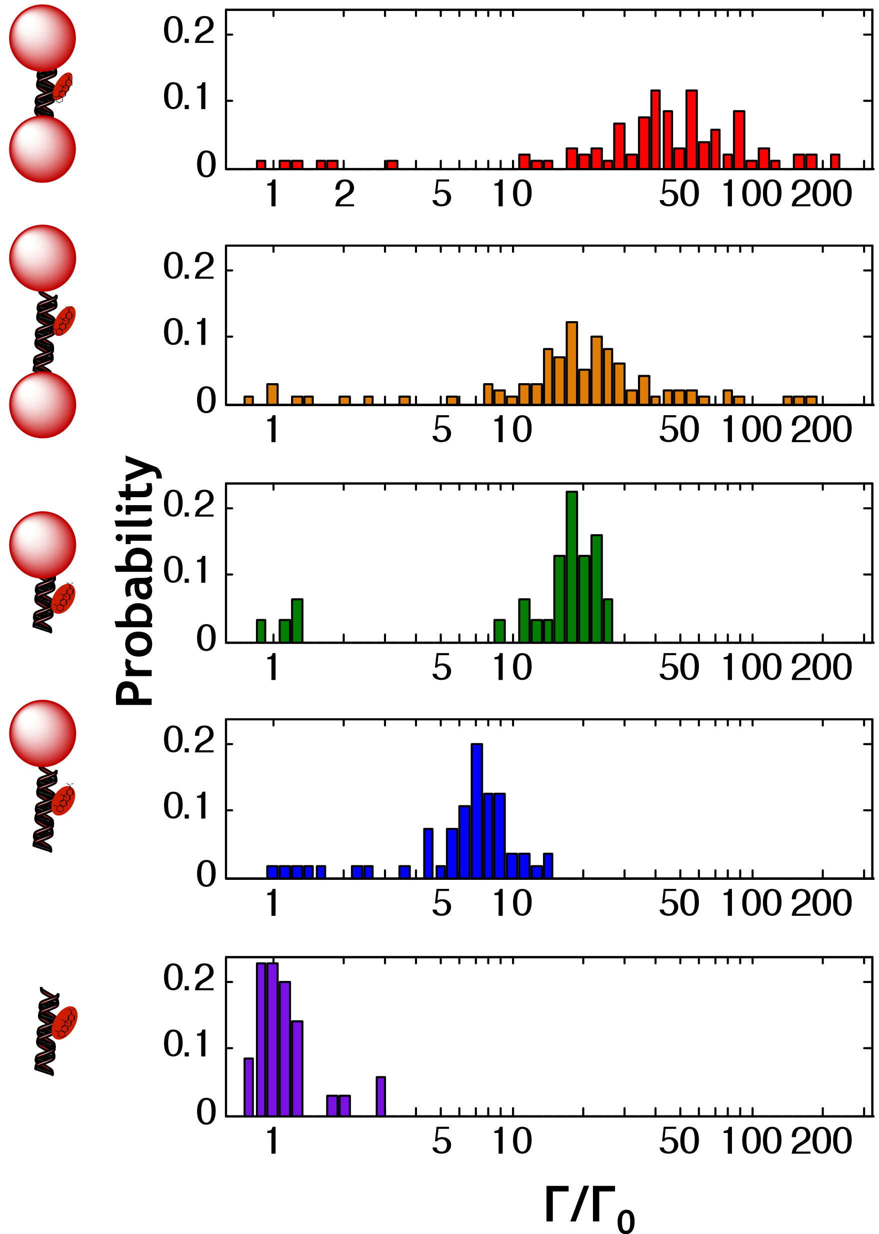
Nanoparticle dimers as optical antennas

Sébastien BIDAULT
(CNRS – ESPCI)

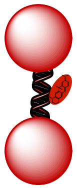




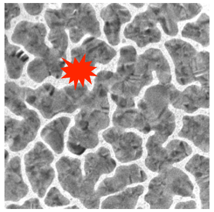




M. P. Busson *et al*,
Nano Lett. (2011)
10.1021/nl2032052



Spontaneous emission in nanostructured environments

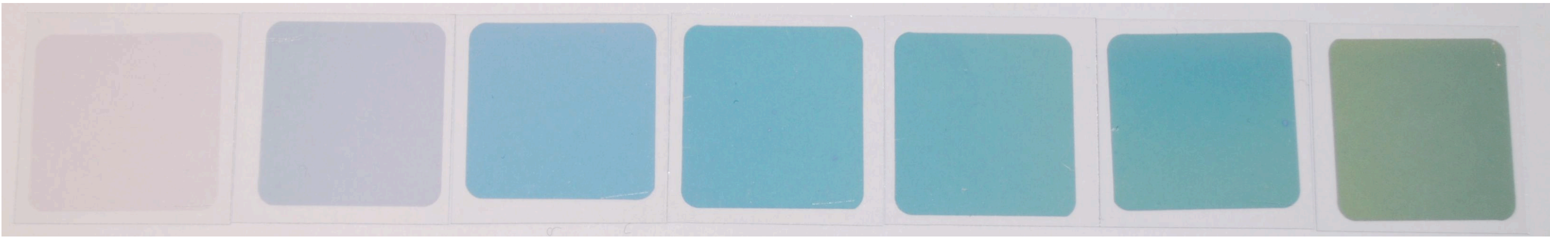


Probing localized plasmons on disordered metallic films

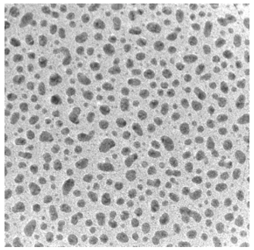
Probing near-field interactions in volume disordered systems (white powders)

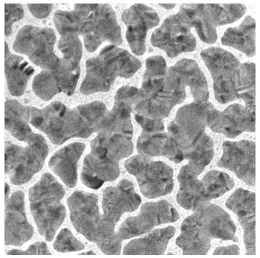
Peculiar optical properties of disordered metal films

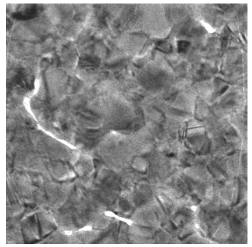
Semi-continuous gold films on a glass substrate



30% **49%** **67%** **79%** **82%** **89%** **99%** Filling fraction





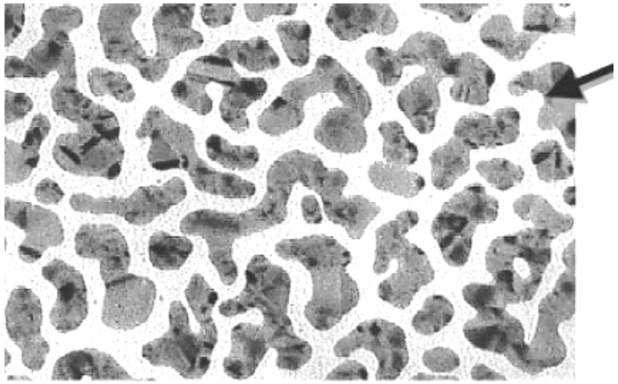


TEM images

P. Gadenne *et al.*, J. Appl. Phys. **66**, 3019 (1989)

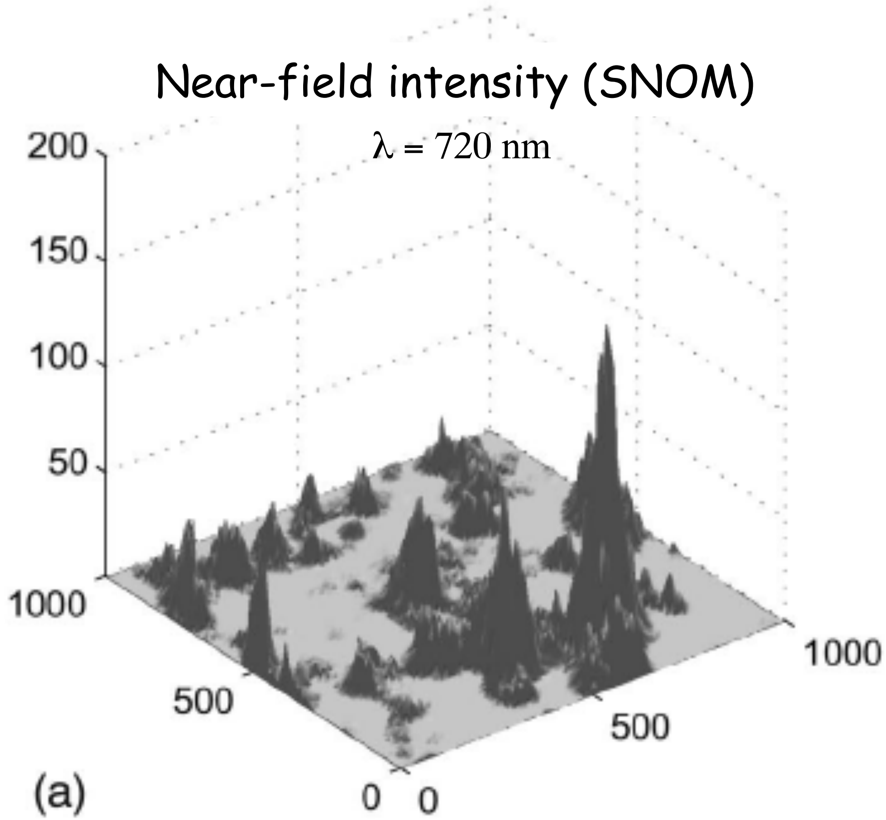
V.M. Shalaev, *Nonlinear Optics of Random Media* (Springer, 2000)

Near-field intensity distribution – « hot spots »



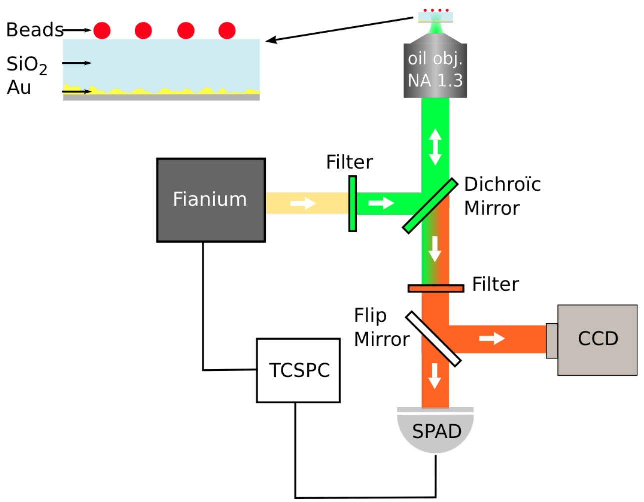
Surface (TEM image)
Gold on glass substrate

Near-field intensity (SNOM)

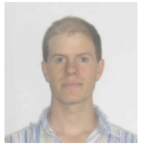


Grésillon *et al.*, Phys. Rev. Lett. **85**, 4520 (1999) ; Phys. Rev. B **64**, 165403 (2001)

LDOS distributions on disordered metal films



V. KRACHMALNICOFF
Post-doc



E. CASTANIE
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Y. DE WILDE
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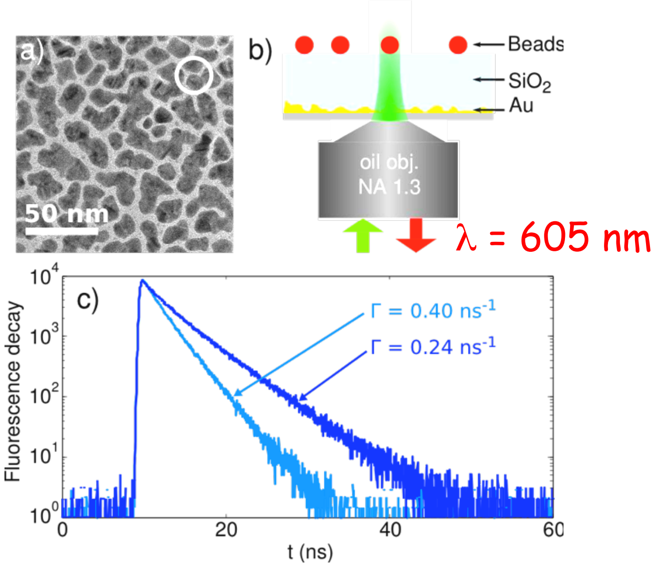
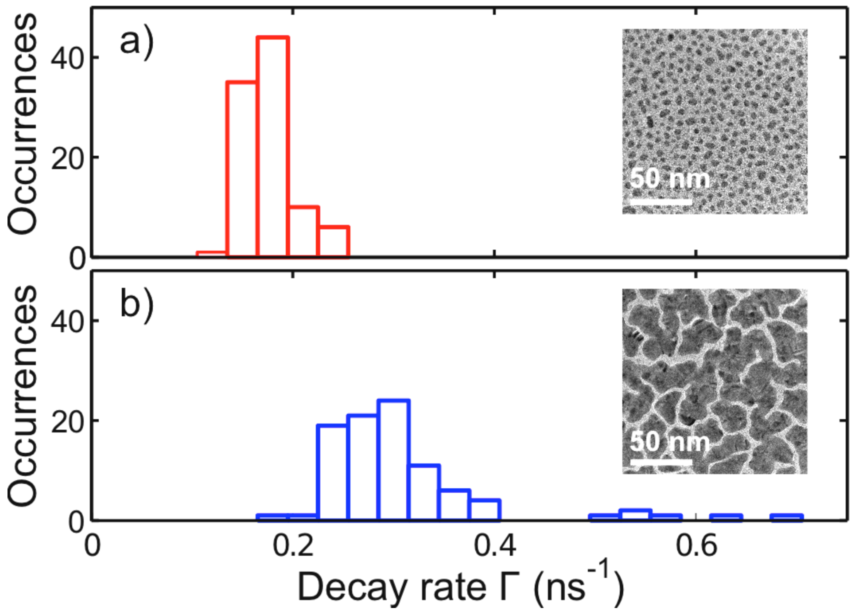


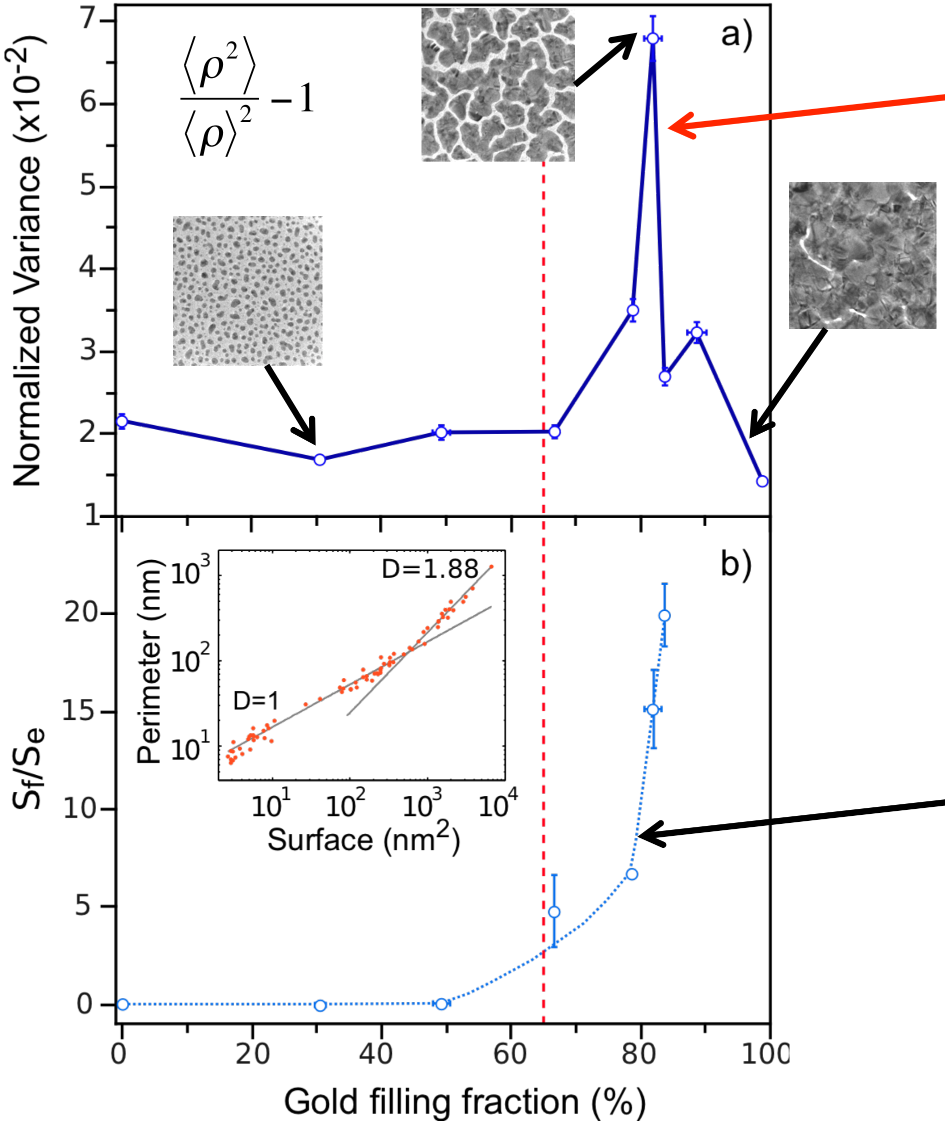
Statistical distributions of Γ (LDOS)



f = 30%

f = 82%

LDOS fluctuations reveals mode localization

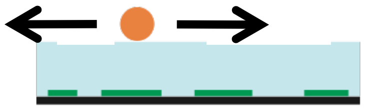


The peak in the LDOS fluctuations is the signature of localized plasmon modes

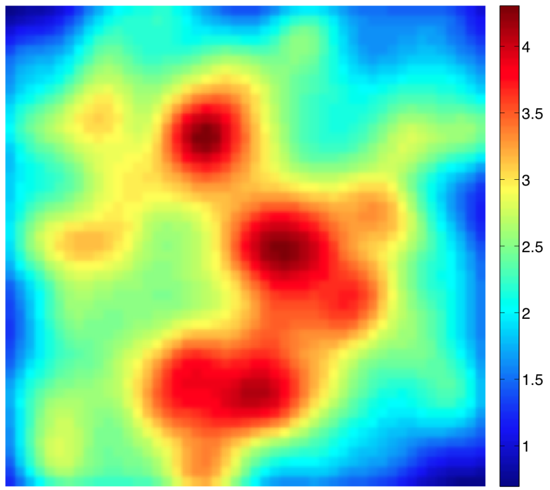
The disordered film is dominated by fractal clusters

Numerical simulation provides additionnal information

$$\Gamma = \Gamma_R + \Gamma_{NR}$$

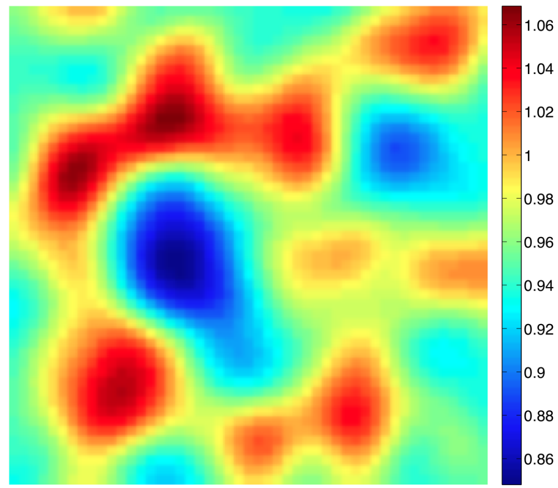


Γ^{NR}/Γ_0

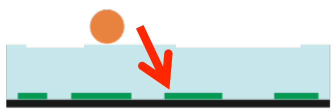


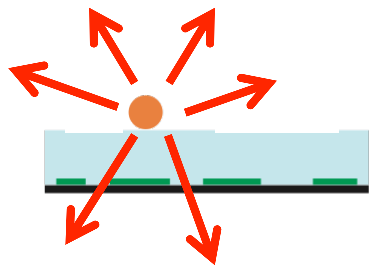
Non-radiative modes

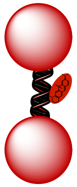
Γ^{R}/Γ_0



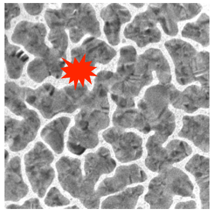
Radiative modes



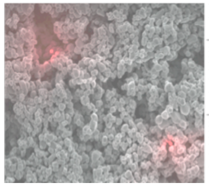




Spontaneous emission in nanostructured environments



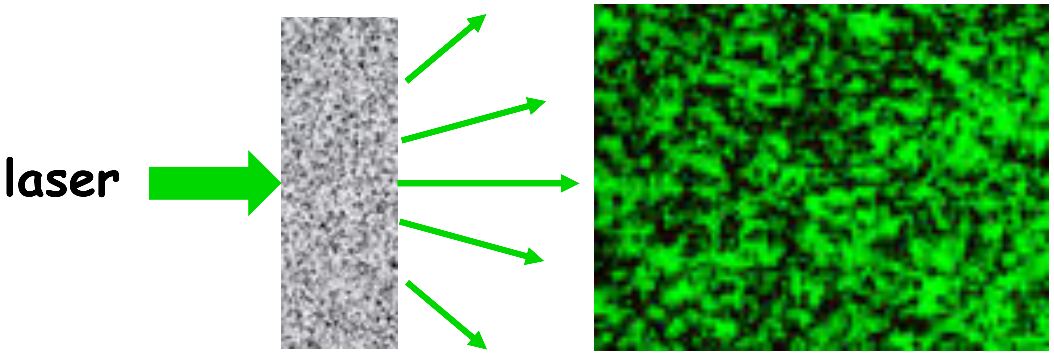
Probing localized plasmons on disordered metallic films



Probing near-field interactions in volume disordered systems (white powders)

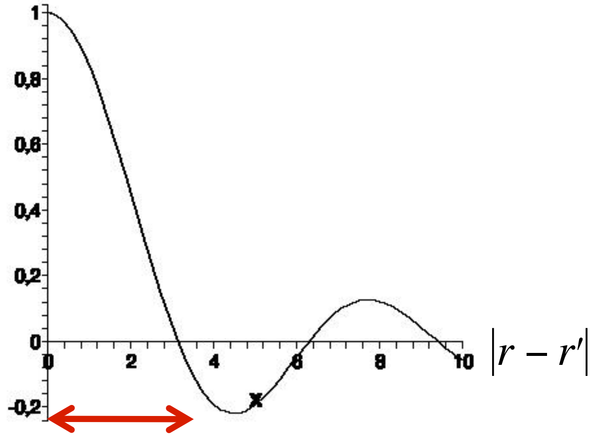
Speckle patterns

Speckle pattern

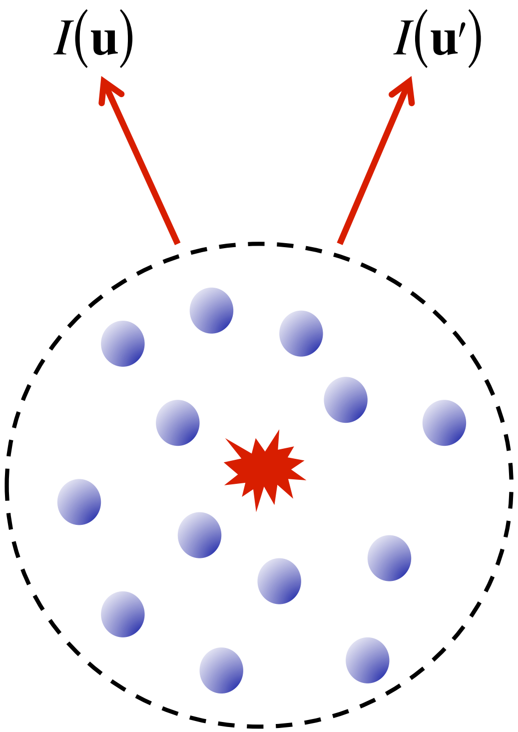


Intensity-intensity correlation

$$\langle I(r)\, I(r')\rangle$$



Speckle produced by a source inside a disordered medium



Infinite-range C_0 speckle correlation

$$C(\mathbf{u},\mathbf{u}') = C_0 + F(\mathbf{u},\mathbf{u}')$$

$$C_0 = \text{LDOS fluctuations}$$

Shapiro, Phys. Rev. Lett. **83**, 4733 (1999)

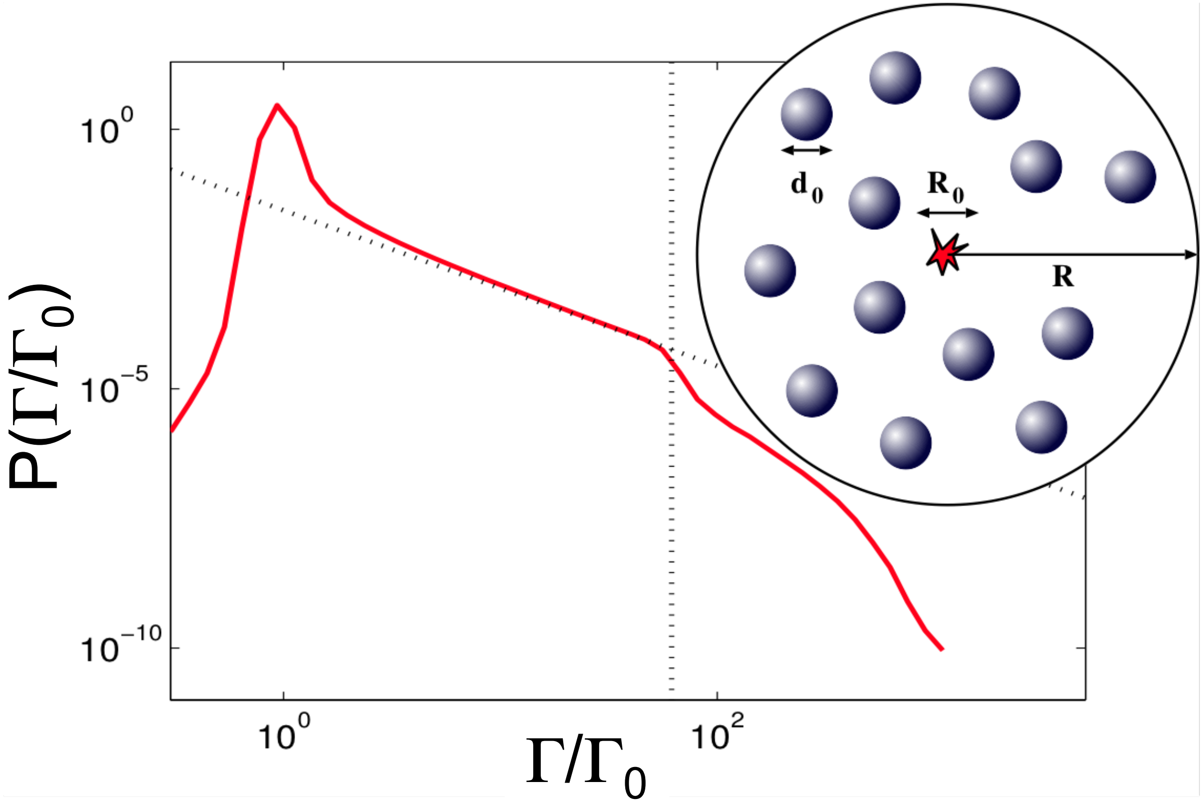
van Tiggelen, Skipetrov, Phys. Rev. E **73**, 045601(R) (2006)

Typical « numerical experiment »

Romain PIERRAT
(CNRS – ESPCI)

Alexandre CAZE
PhD student

Statistical distribution of decay rate Γ (LDOS)

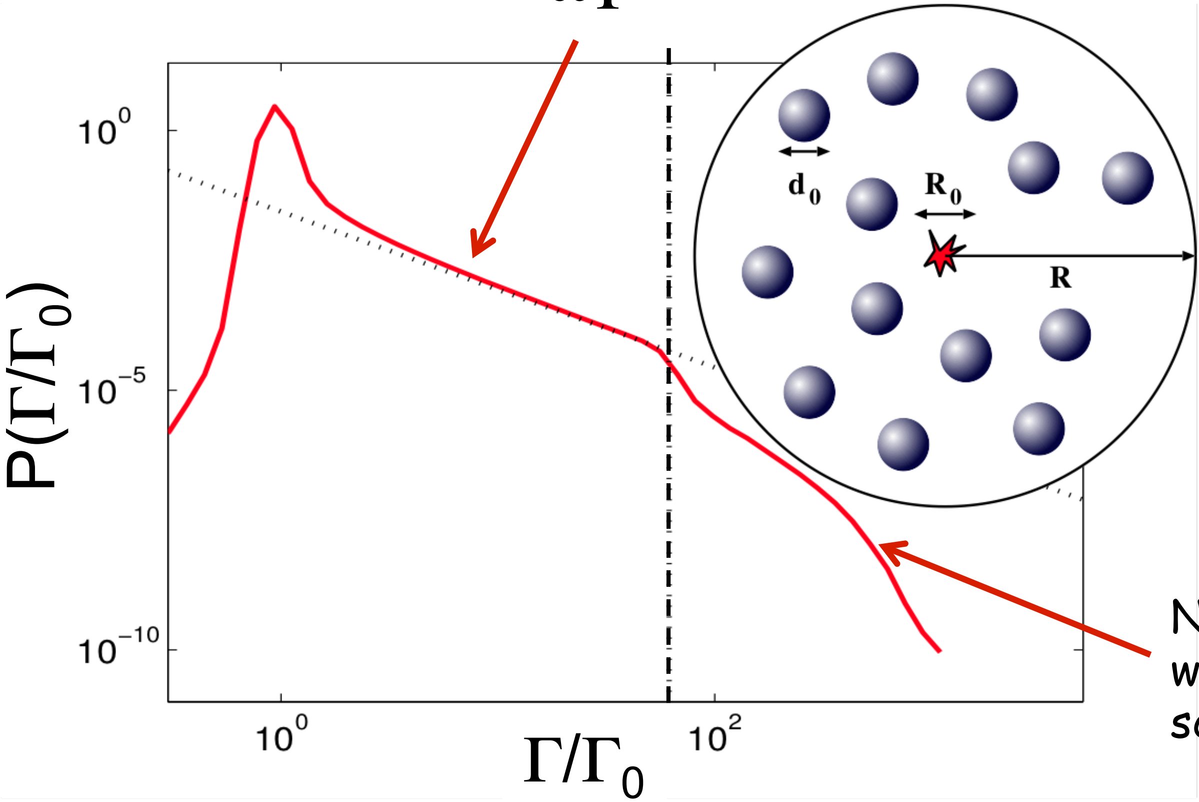


- Resonant point scatterers (« atoms »)
- $\lambda \approx 630$ nm
- Cluster size R = 1.2 μm
- Exclusion volume R_0 = 50 nm

Long tail: Near-field interactions

Near-field interaction
with <u>one</u> scatterer

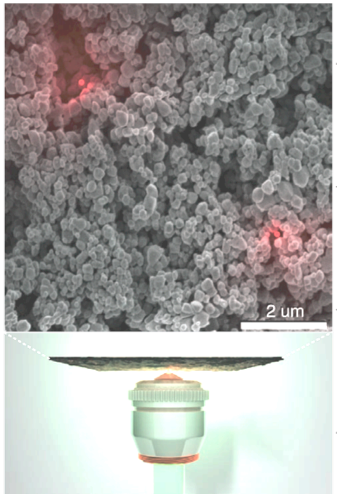
$\propto \Gamma^{-3/2}$



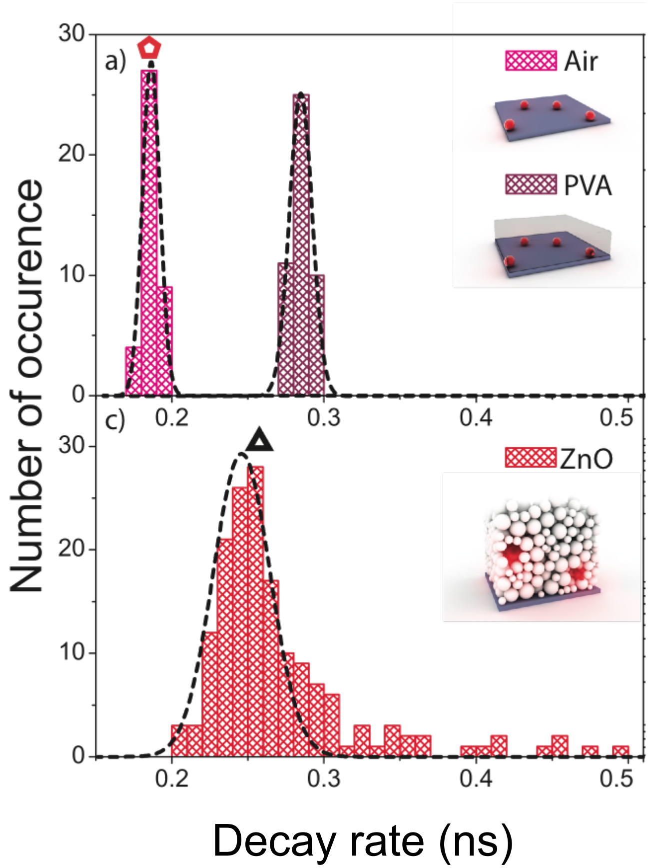
Near-field interaction
with <u>more than one</u>
scatterer

Broad - asymmetric distribution of LDOS (Purcell factor)

Riccardo SAPIENZA
Niek van HULST
(ICFO Barcelona, Spain)



LDOS statistics probed by lifetime
of nanosources (24 nm fluorescent beads)



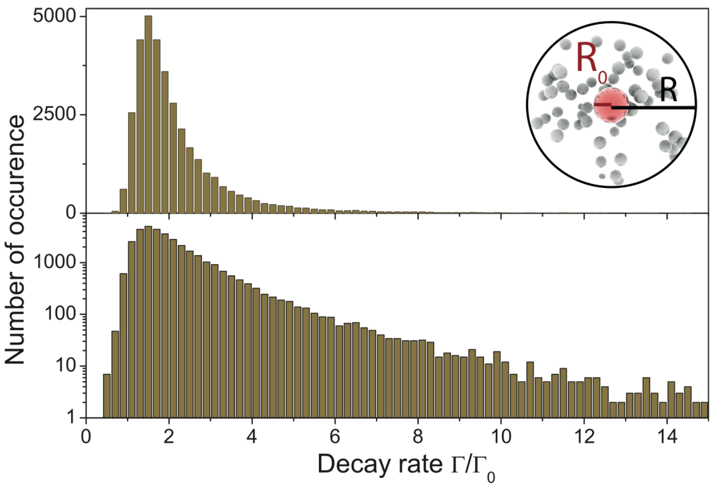
ZnO powder
Polydisperse particles
$(140 \pm 50\ nm)$

Photon mean free path

$$\ell = 0.9\ \mu m$$

$$k\ell = 9.4$$

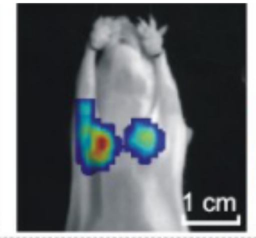
Theory



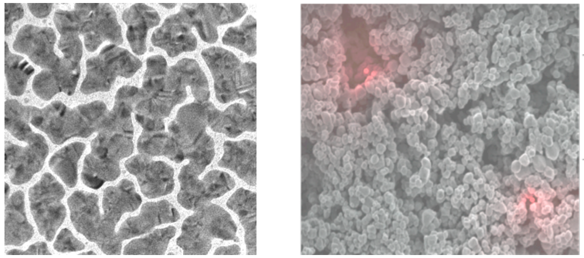
Sapienza, Bondareff, Habert, Pierrat, Carminati, van Hulst, PRL **106**, 163902 (2011)

Coupling spontaneous emission with disorder: Why ?

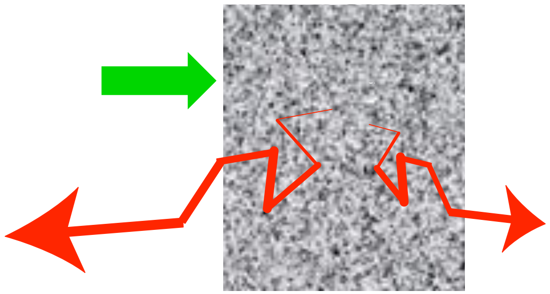
Fluorescence imaging
in complex media



Nanophotonics - Novel materials



Novel light sources
(e.g. random lasers)



Fundamental studies of light
transport in scattering media
(e.g. probing Anderson localization)